A CLASSIFICATION OF EXTRATERRESTRIAL SPHERULES FOUND IN SEDIMENTARY ROCKS AND TILL[1]

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ABSTRACT

Extraterrestrial spherules have been recovered from Pleistocene drift and from more ancient sedimentary rocks. Modern deposits also contain spherules, but, because a significant percentage of these spherules can be shown to be of recent terrestrial and industrial origin, such deposits are not dependable sources of extraterrestrial spherules.

A classification of extraterrestrial spherules has been developed, based on microscopic observation, which shows a continuous series between two end members. One end member is magnetic and has a luster that varies from bright metallic to black submetallic, probably indicating a composition varying between iron and magnetite. The surface is notably reticulated. The other end member is nonmagnetic and is a generally colorless to amber-colored, transparent glass, probably with a composition varying between those of olivine and pyroxene. The surface is usually smooth, though submicron-sized "percussion" marks or micron-sized indentations may be observed on some specimens. These members are referred to as Type I and Type III, respectively. Spherules intermediate between these end members are called Type II.

Recent articles dealing with extraterrestrial spherules in ancient sediments (Thiel and Schmidt, 1961; Skolnick, 1961; Hunter and Parker, 1961; Crozier, 1960), and in shallow firn and deep ice cores from Greenland (Langway and Marvin, 1964), in general discuss "black magnetic spherules," but there is little or no description of silicate spherules. There has been no comprehensive classification of the various types of spherules, although Schmidt et al. (1963) describe three types of "metallic" spherules, based on surface features. The present paper offers data supporting the view that ancient spherules have a continuous range in composition from mainly iron and/or magnetite, with a metallic luster, to magnesium-iron silicate, with a glassy luster.

MODERN EXTRATERRESTRIAL DUST AND SPHERULES

Considering first the nature of the present accretion of extraterrestrial material by the earth, Hemenway and Soberman (1962), from "studies of micrometeorites obtained from a recoverable sounding rocket," report particles as falling into three types: "high density spheres, medium density irregular particles, and extremely irregular medium density particles (fluffy particles)." Dubin and McCracken (1962), from data obtained by the use of satellites and space probes, conclude that there is an accretionary rate of about 10^4 tons per day (3.65×10^6 tons per year). Pettersson (1960) collected dust from two sites in the Hawaiian Islands, one at 11,000 feet and the other at 10,000 feet altitude; using a nickel content of 2.5% as an average for all meteorites, he calculated the infall of extraterrestrial dust to be 14×10^6 tons per year, a figure later modified to 5×10^6 tons per year. Alexander, McCracken, and LaGow (1961) report on data recorded by 1959 Eta (Vanguard III) that "the impacts of approximately 2800 dust particles with momenta greater than 1×10^{-2} dyne sec. were recorded during the 70-hour interval, which coincides closely with the time of occurrence of the Leonid meteor shower. This number of impacts is approximately equal to the number of impacts observed during the remainder of the lifetime (approximately 78 days) of the experiment." It is, therefore, possible that Dubin and McCracken's figure could be raised by the addition of periodic increased infall of dust attending meteor showers. Mutch (1964) reports quite tentative rates of accumulation for three samples of Silurian salt collected from the Cayuga Rock Salt Mine of 1.4×10^8,

[1]Manuscript received October 30, 1964.

3×10^8, and 10^9 metric tons. He states that these samples contain a high percentage of spherical and subspherical particles. He says, "It can be argued that a large proportion, if not the majority, of the particles weighed is of terrestrial origin," and concludes, "The strong suggestion remains that extraterrestrial additions to the earth 400 million years ago were as great as they are at the present and possibly much greater."

Considering spherules only Crozier (1962) reports, from a five-year continuous collection made at a station located in central New Mexico, an annual accretion rate of 1.6×10^5 metric tons of "black magnetic spherules." This rate is based on spherules ranging in size from 5 microns up to 100 microns in diameter. However,

TABLE 1
Frequency Distribution of the three types of Spherules counted on selected samples

<table>
<tr><th>Specimen</th><th>Description</th><th>Type I</th><th>Type II</th><th>Type III</th></tr>
<tr><td>1</td><td>Pennsylvanian coal
−200 mesh, ignited 700°F for
24 hours</td><td>27[a]</td><td>20[b]</td><td>11</td></tr>
<tr><td>2</td><td>Pennsylvanian coal
−200 mesh, ignited 650°F for
18 hours</td><td>90[c]</td><td>73</td><td>4</td></tr>
<tr><td>3</td><td>Moraine, Nebraskan
−200 mesh</td><td colspan="2">|———10———|</td><td>6</td></tr>
<tr><td>4</td><td>Marl
8000–9000 B.P. (Forsyth '61)
−100 mesh</td><td>74</td><td>63</td><td>1[d]</td></tr>
<tr><td>5</td><td>Navajo sandstone
Lower Jurassic</td><td>9</td><td>21</td><td>7</td></tr>
<tr><td>6</td><td>Wingate sandstone
Lower Jurassic</td><td>4</td><td>15</td><td>2</td></tr>
</table>

[a]Includes 6 red magnetic spherules.
[b]3 irridescent spherules were observed.
[c]Includes 10 red magnetic spherules.
[d]Probably most of small silicate spherules lost in preparation, (decanting clay fraction). 2 irridescent spherules observed. 15 type II smooth spherules observed.

TABLE 2
Size Distribution of spherules in specimen 2, table 1, in microns

10	10–20	20–30	30–40	40–50	50–60
<36	60	47	16	4	4

this rate for the annual accretion of spherules larger than 15 microns is only 0.5×10^5 tons. Thiel and Schmidt (1961) studied spherules contained in Antarctic ice cores, but only "black" spherules were used by them to calculate an accretionary rate of 1.84×10^5 tons per year. They did report "some yellow or brown spherules, a number of which are translucent," but did not include them in their estimate. Their count was further restricted to spherules over 15 microns in diameter. As Crozier (1962) points out, "There is thus disagreement by a factor of 3.7 between the two determinations, for the common size interval."

As pointed out by Handy and Davidson (1953), and Hodge and Wildt (1958), the problem of collecting extraterrestrial spherules presently falling onto earth is that they are for the most part indistinguishable from spherules associated with industrial contamination. From a population of 32 spherules collected in the desert near San Diego, California, Fredriksson and Gowdy (1963) determined the "cosmic" origin of two spherules by their nickel-iron content, using an electron microprobe to scan polished sections. They considered the other thirty iron-oxide spherules to be artificial contamination from San Diego. Spherules asso-

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EXPLANATION OF PLATE I

FIGURE 1. Type I. Black magnetic spherule—lower spherule. Note surface reticulation. Upper spherule is Type II black smooth magnetic spherule. Pittsburgh No. 8 coal seam, Ohio. Pennsylvanian.

FIGURE 2. Type I. Dark gray metallic spherules, magnetic, with distinct surface reticulation. From Pennyslvanian Pittsburgh No. 8 coal seam, Ohio.

FIGURE 3. Type I. Black magnetic spherule with reticulated surface showing similarity of surface to the surface on surrounding magnetite grains. Till of Defiance Moraine. Ohio. Late Woodfordian, about 15,000 B.P. (Frye and Willman, 1963).

FIGURE 4. Type I. Red non-magnetic spherule, probably hematite or hydrous iron oxide, possibly originally iron or magnetite, or both. From ice-contact stratified drift just south of Buchanan, Indiana. Probably of comparable age to the Valparaiso Moraine. Late Woodfordian substage, about 15,000 years B.P. (Frye and Willman, 1960).

FIGURE 5. Type II. Smooth black magnetic spherule, appears slightly transparent peripherally, with a suggestion of a large central dark irregular inclusion. From Wingate sandstone, Zion National Park, Utah. Lower Jurassic.

FIGURE 6. A cluster of spherules fused together. The two smaller spherules are Type I, dark metallic with reticulated surface. The large spherule is about ⅓ Type I where it is fused to the middle spherule, whereas the other ⅔ of the surface is Type II smooth. Specimen from an Illinoian kame, Highland County, Ohio. Compare to Figs. 14, 22, and 28.

FIGURE 7. Type I. Gray metallic with a bit of matrix clinging to it—upper spherule. Lower spherule Type II black; no reticulation, but with probably numerous submicron "percussion" pits on the surface reducing reflectivity. From marl deposits north of Castalia, Ohio. 8,513±500 years B.P. (Forsyth, 1961).

FIGURE 8. Type I. Gray metallic reticulated group clinging together by magnetic attraction after collection with an alnico magnet. From marl deposits north of Castalia, Ohio. 8,513±500 years B.P. (Forsyth, 1961).

FIGURE 9. Type I. Dark gray metallic reticulated in three directions (East, North, and West with a smooth black Type II spherule in central location. From marl deposits north of Castalia, Ohio. 8,513±500 years B.P. (Forsyth, 1961).

FIGURE 10. Type I. Metallic gray reticulated spherules clinging to irregular grains of magnetite after collection with alnico magnet. From Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.

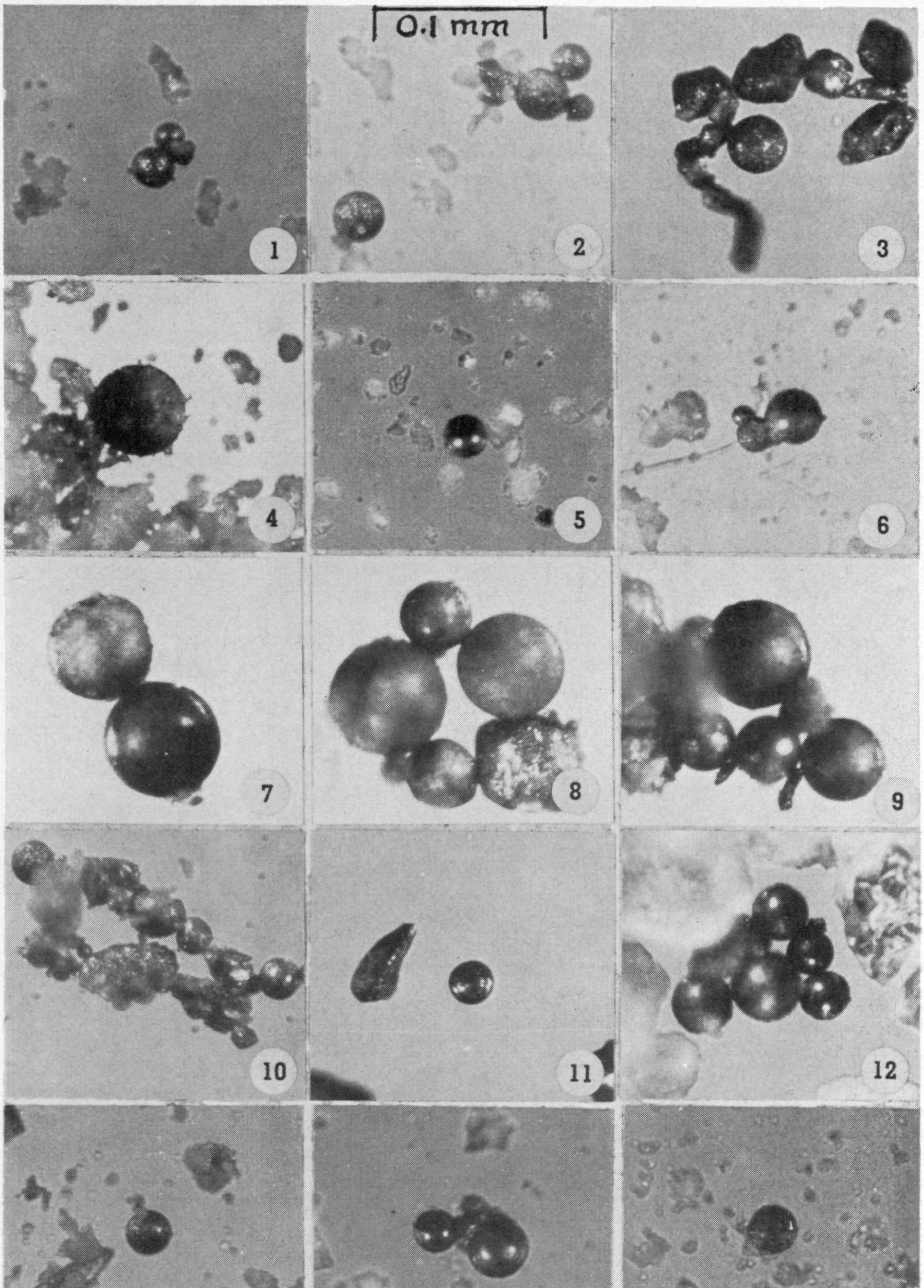
FIGURE 11. Type I. Brown magnetic reticulated spherule. Probably hydrated iron oxide on the outside, but presenting a surface of high reflectivity. Till of Defiance Moraine, Ohio. Late Woodfordian, about 15,000 years B.P. (Frye and Willman, 1963).

FIGURE 12. Two large spherules are Type I dark gray metallic and reticulated. The three smaller spherules are Type II. The lower luster of the two larger spherules, is probably due to the variation in concentration of submicron pits. All magnetic. From marl deposits north of Castalia, Ohio. 8,513±500 years B.P. (Forsyth, 1961).

FIGURE 13. Type II. Black magnetic spherule with an obvious concentration of "percussion" pits dulling the luster. From Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.

FIGURE 14. Fused cluster. Large sperhule is a Type I where fused to irregular metallic grain, but the free part is Type II pitted. The spherule on the left is also fused to the same irregular grain and is Type II black, apparently with a black transparent exterior enclosing an irregular opaque grain. The upper small spherule which is indistinct in the picture owing to limited depth of focus is a Type III spherule of brown opaque glass. I suggest that the black irregular reticulated magnetite grain diffused onto the large right sphere which was originally (probably of extremely short duration) a typical Type II. Navajo sandstone, Zion National Park, Utah. Lower Jurassic.

FIGURE 15. Type I. Dark gray metallic reticulated spherule which appears to have a patch of surface on the upper right side similar to a Type II spherule. Compare with Fig. 6, large spherule; Fig. 14, large spherule; Fig. 23; and Fig. 24. From Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.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ciated with a meteor fall (Krinov, 1960) attest to their extraterrestrial origin, but with what confidence can one state that randomly collected suites do not contain spherules from industrial contamination? Fredriksson and Martin (1963) point out volcanic activity as a further possible source of contamination, but this would seem to be quite restricted in time and place. Therefore, because modern industrial contamination is certainly real and widespread, I have restricted this study to ancient populations of extraterrestrial spherules.

ANCIENT EXTRATERRESTRIAL SPHERULES

The advantage in studying ancient extraterrestrial spherules is that the problem of industrial contamination is eliminated; however, there still remains the problem

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EXPLANATION OF PLATE II

FIGURE 16. Type II–III. Clear colorless glass magnetic spherule with a large irregular magnetic fragment enclosed. Photo overexposed. Specimen from Green River Shale, Eocene.

FIGURE 17. Type II. Typical smooth black clear glass magnetic spherule with a large irregular magnetic fragment enclosed. From Illinoian kame, Highland County, Ohio.

FIGURE 18. Type II. Dark brown smooth spherule with "percussion" pits, slightly magnetic, not a common variety. Wingate sandstone, Zion National Park, Utah. Lower Jurassic.

FIGURE 19. Type II. Typical smooth black magnetic spherule. From Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.

FIGURE 20. Type I–II. High-reflection black magnetic spherule with a reticulated network on the surface. From Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.

FIGURE 21. Type II. Rather large, typically black, smooth high-reflecting spherule; irregularities due to minute particles of matrix clinging to it. Wingate sandstone, Zion tional Park, Utah. Lower Jurassic.

FIGURE 22. Type III. A rather remarkable cluster of transparent colorless Type III spherules fused together. Impossible to get all of them in focus. The large left-side spherule is a thin-walled bubble. The large out-of-focus central spherule is almost solid, but has small bubble inclusions. The smaller upper-right-hand spherule is solid, but has a small dimple. Navajo sandstone, Zion National Park, Utah. Lower Jurassic.

FIGURE 23. Type III–II. Translucent colorless glass, Type III for the most part, but with a small Type II cap on the left side. A transitional type between Type III and Type II. The entire surface appears to have submicron- to micron-sized dimples. I do not know whether dimpling is the result of the preparation or not. The sample was held at 700°F for 24 hours to eliminate organic material. Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.

FIGURE 24. Type II–III. Transitional type (cf. Fig. 23). Left side of specimen is Type II smooth black spherule, presumably magnetic. The right side is Type III transparent colorless glass. Specimen from Navajo sandstone, Zion National Park, Utah. Lower Jurassic.

FIGURE 25. Type III. A thick-walled bubble of transparent colorless glass. Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.

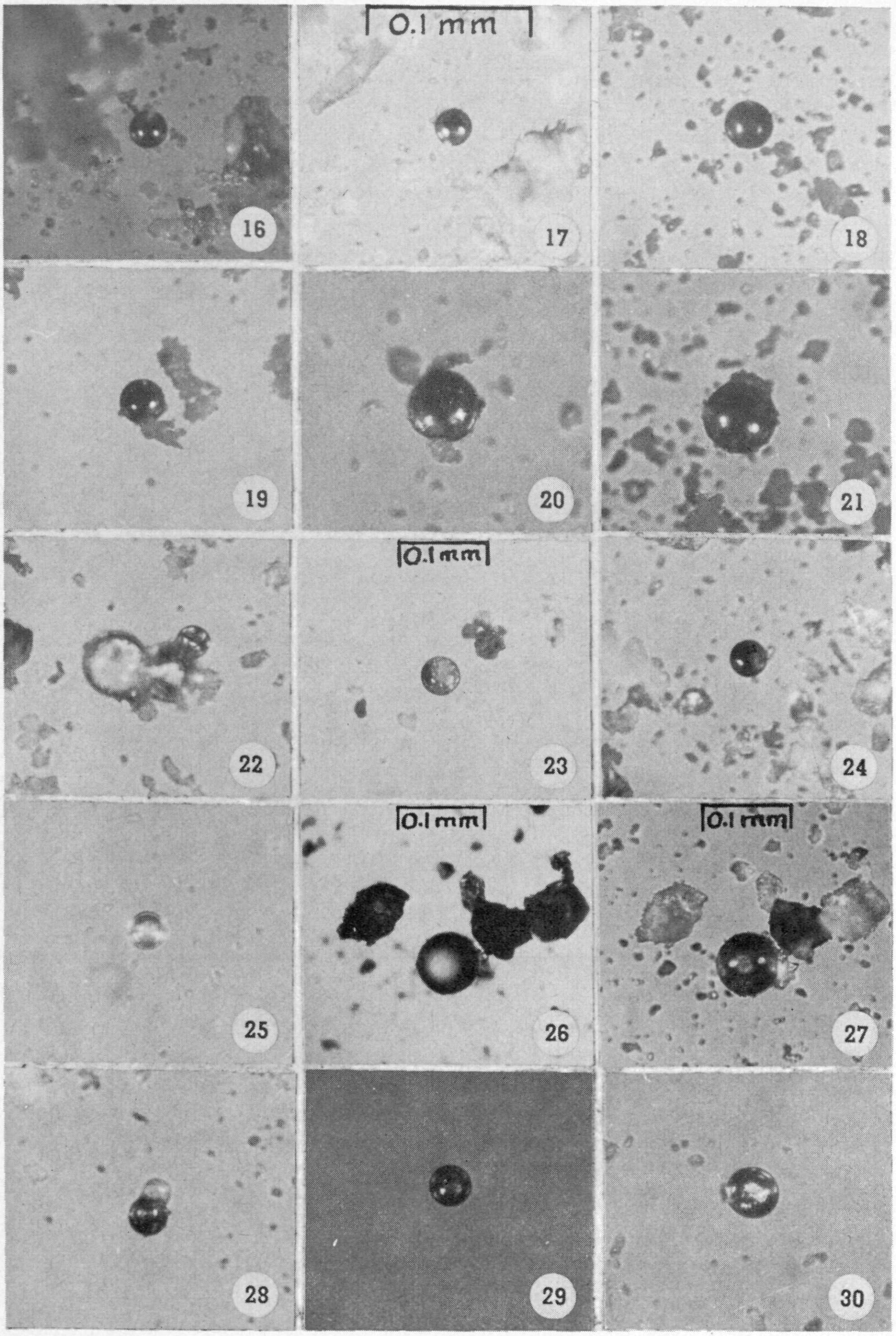
FIGURE 26. Type III. Solid smooth transparent colorless glass spherule with substage convergent light showing light transmitted through it. Brown "deltaic" sand from quarry at Este Ave., Cincinnati, Ohio, probably early Illinoian. (Personal communication R. H. Durrell, University of Cincinnati, 1961).

FIGURE 27. Type III. Same specimen as Fig. 26, but with parallel substage lighting plus overhead lighting. Note that these solid transparent glass spherules could, on cursory examination, be mistaken for Type I or Type II spherules. They often contain minute irregular grains of magnetic material, presumably magnetite or iron which carries them into the magnetic recovery fraction. Brown "deltaic" sand from quarry at Este Ave., Cincinnati, Ohio, probably early Illinoian. (Personal communication R. H. Durrell, University of Cincinnati, 1961).

FIGURE 28. Types II and III fused together. From Navajo sandstone, Zion National Park, Utah. Lower Jurassic. Cf. Figs. 6, 14, and 22.

FIGURE 29. Type III. Transparent amber-colored glass spherule with a centrally located bubble inclusion about half the radius of the spherule. Specimen from Valparaiso Moraine, Indiana, about 1.3 miles south of location of specimen in Fig. 4.

FIGURE 30. Type III. Transparent brown colored glass spherule from Pennsylvanian Pittsburgh No. 8 coal seam, Ohio.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of volcanic contamination. Fredriksson and Martin (1963) describe "black spherules" found in Pacific islands as "usually thin, empty and fragile shells." They state that "although they are very similar in external appearance to cosmic spherules and to artifacts, their composition and structure are markedly different." They say further:

> The discrepancy between values for sediments and ice is possibly due to the crudity of methods employed for extracting spherules from sediments, in that the extraction tends to destroy the fragile volcanic spherules.

I assume that they are referring, in part at least, to the findings of Pettersson (1960), in which, on the basis of cores from the Mediterranean, he calculated an annual accretionary rate of 3,300 tons per year. This compares to Thiel and Schmidt's (1961) estimate of 184,000 tons per year, based on cores from Antarctic ice. It is their assumption of the wide distribution in time and place of such volcanic spherules that is probably open to question. Buddhue (1950) examined samples of volcanic dust, and reported: "No silicate or other spheres were found." I have examined ash from Mt. Lassen, but ten hours of search failed to reveal one spherule. Of notable import, too, is the finding of Hodge and Wildt (1958), collecting spherules in New Haven, Connecticut, who state, "That they are predominantly industrial in origin is apparent from the fact that their number drops by a factor of 2 on Sundays, and by a factor of 4 at 10 miles from the center of the city." This observation points out the marked falloff of industrial spherules in a population collected at localities of increasing distance from the source, a generalization that may also apply to volcanic sources. It is questionable, therefore, whether suites of spherules collected at any great distance from volcanic activity would be contaminated appreciably, if at all. But, more than that, volcanic activity is highly sporadic in time and place, and it would appear fortuitous indeed to find volcanic contamination in any ancient sediments, with the possible exception of the few which could be assigned a direct relationship in time and place with active volcanism. I spent a considerable length of time searching for spherules in beach sands, including specimens collected in California, Oregon, and Hawaii, and all samples appeared to have similar populations of spherules. Relatively few spherules were found per unit volume of beach sand, compared to sand from other, less active depositional sites. However, the Hawaiian sands had no increase of spherules compared to the others, which probably corroborates the view of Fredriksson and Martin (1963) that volcanic spherules are very fragile. I have also noted the great number of hollow black magnetic spherules and flask-shaped spherules close to industrial sources, but have been impressed with the rapid fall-off of these types in collections made only a few miles distant from the source. Because of these several observations, I consider contamination by volcanic activity to be limited to sediments near the place of origin, occurring only where rapid isolation and preservation of the spherules are possible, and to be greatly restricted stratigraphically. For these reasons, I believe that the spherules discussed in the following study are exclusively extraterrestrial.

Crozier (1960) treats only black magnetic spherules obtained from ancient shales and carbonate rock. This present paper deals with all spherules obtained from a variety of ancient sedimentary rocks: aeolian sandstone, coal, till, marl, and one shale.

Apparently some spherules are formed by ablation of meteors (Krinov, 1960). It may be that most of them are formed during numerous subvisual meteor showers. Gallagher and Eschelman (1960), reporting on observations by radar of ionized meteor trails as small as the 15th visual magnitude, say, "This leads to the estimation that there must be millions of shower orbits which intersect the orbit of the earth." They conclude, "Thus the observed particle groupings in the meteor population may indicate the presence of a large number of small, subvisual comets." Considering the great concentration of interplanetary dust particles associated with the Leonid meteor stream (Alexander et al., 1961), it would seem reasonable to assume a similar concentration of dust in the subvisual comets. It would further seem reasonable to assume that some of the perigees of these dust concentrations, or subvisual comets, would be in a solar proximity that would result in spherule formation of at least some of the dust particles. Turbulence of rapidly expanding gases (as in coma formation) may cause impact and abrasion of the solid spherules at a later stage, producing the "percussion" pits observed on the surface of some varieties of spherules mentioned later in this paper. I agree with Schmidt et al. (1963) that it is unlikely that the surface features of spherules are due to terrestrial processes; however, it has not as yet been shown that turbulence during shower activity in the upper atmosphere could not be held accountable.

PROCEDURE

Specimens of the sedimentary rocks studied were carefully trimmed to remove surface contamination, then pulverized, and sieved. Most spherule populations occurred in the -200-mesh fraction (-74 microns) and only occasionally in the coarser fractions studied.

Black magnetic spherules may be recovered with an alnico magnet covered with saran wrap, but recovery of glassy (silicate) spherules presents a difficult problem. If the glassy spherule has minute magnetic inclusions, they may be recovered by very gentle manipulation of the magnet. If, however, they are nonmagnetic (and this is generally true for the glassy spherules that are less than 20 microns in diameter), a sample of the whole fraction may be dusted onto a petrographic microslide and then traversed by the use of a mechanical stage mounted on the microscope. This search is extremely time consuming because of the dilution factor, but is materially aided by use of top and bottom lighting, together with a binocular petrographic microscope using 100 power.

To eliminate the clay fraction of a specimen, careful decanting of an alcohol or acetone suspension was used with some success, but, because of their low specific gravity, glassy spherules are easily decanted too, especially if they contain bubbles. Much transferring of the dried sample results in a loss of some of the -20-micron-size glassy spherules, because, owing to static electricity, they tend to cling to dry surfaces. Several other methods were tried, including centrifuging in heavy liquids, but the recovery of glassy spherules was greatest when the handling was at a minimum. Over 500 photographs were taken, using Panatomic X and Professional Kodachrome II films. Occasionally a barren control specimen was run in conjunction with a sample to check for laboratory contamination. None was observed. However, because cover slips were not used, since they interfered with top lighting, there was always the possibility of contamination; I see no way to assess this factor at this time. Where necessary, prepared microslides were ignited for several hours to eliminate organic material, including spores and pollen, which in some cases might have been mistaken for spherules. Subsequent microscopic observation revealed that the numerous variations of spherules could be described by a single three-fold classification, so counts were made on a few of the specimens to gauge the distribution of the three types.

CLASSIFICATION OF SPHERULES

Despite the numerous variations of spherules observed, all appeared to fit into a three-fold classification. Each of the three general types included in this classification are described below. *Type I:* Metallic grey to submetallic black, magnetic. Some specimens are bright red, others are reddish brown or dull brown, all colors considered to be due to oxidation. Other spherules are mottled black and red, or they may be black on one side and red, brown, or white on the other. Occasionally they occur fused to Type-II or Type-III spherules. The size ranges from less than 5 microns to over 1,000 microns in diameter, the average falling between 10 and 30 microns; this average might be lower if it were not for the fact that sizes smaller than 10 microns are difficult to separate out. The larger spherules may occur in the form of flasks or hollow spheres. The surface always appears reticulated, with micron- to submicron-sized grooves and ridges. The composition in most cases is probably magnetite, but may be essentially meteoritic iron or a metallic core with a magnetite crust, or a hematitic or hydrous iron oxide crust. Grades into Type II.

Type II: Black shiny opaque, often smooth surfaced, but generally covered with what appears to be submicron-sized "percussion" marks; may have a reticulated surface; may also be slightly puckered, or have many small indentations; occasionally hollow or flask-shaped. Most Type II spherules are magnetic to slightly magnetic, and are rarely unresponsive to an alnico magnet. Their size range is similar to that of Type I, though the average is usually a little less, that is, from 10 microns to 25 microns. The surface material ranges from black to dark brown, to yellow, to colorless. The darker material is opaque and the yellow to colorless material is transparent. Spherules with transparent glassy surfaces contain irregular black magnetic cores. In this case, one large fragment may occupy almost the whole interior of the spherule, or there may be one or several small irregular blebs eccentrically arranged within the spherule. It is impossible, in some cases, to determine whether the spherule should be classed as Type I or Type II; such cases are referred to as Type I–II. For this reason, it is probably correct to conclude that the two types are gradational.

Type III: Glassy, usually transparent colorless, but may be amber (fairly common), yellow, brown, or red (rare). A few are translucent, or opaque white. The size range is from less than 5 microns to over 600 microns in diameter; the average falls between 15 and 30 microns. Many of the colorless transparent varieties contain bubble inclusions; the presence of abundant submicron-sized bubbles probably explains the translucent white appearance of some spherules. Numerous spherules in the size range from 20 to 40 microns are simply hollow glass bubbles, whereas other spherules contain one or more bubble inclusions with random distribution. Generally the smaller spherules (those less than 15 microns in diameter) appear to be composed of solid, transparent glass, and are isotropic (with crossed polars). Many of the glassy spherules of all colors contain black opaque inclusions and, because of their magnetic response, these are presumed to be magnetite or iron. When the size of an inclusion is large with respect to the size of the spherule, it is difficult to call it either Type II or Type III, and then it is referred to as Type II–III. Because of this relationship it is believed that Type II spherules are gradational into Type III.

From the above observations, I suggest that extraterrestrial spherules may be classified into three types, I, II, III, and that the three types form a continuous series. An extended generalization may be that there are actually only two types of material dominating the composition of the spherules, one being magnetic material, magnetite and/or meteoritic iron, the other being glassy with a composition similar to olivine and/or pyroxene. In this case, the three-fold classification would represent intervals of the range between the two basic types of material. In most examples, a spherule could readily be assigned to one of these three types. Occasionally, when doubt demanded it, an intermediate classification could be used, such as Type I–II or Type II–III.

ACKNOWLEDGMENTS

I am most appreciative of the funds over the past two years from the University Fund for Research Aid, Bowling Green State University, for the purchase of equipment and photographic supplies which made this study possible. I am indebted to R. D. Hoare for helpful discussion and to J. R. Coash for two of his specimens of till.

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